



Anodic polarization behaviour of cast iron alloys in deaerated concentrated H_3PO_4 solutions containing Cl^- and F^- ions

M.S. ABDEL-AAL and A.A. HERMAS

Chemistry Department, Faculty of Science, Assiut University, Assiut, Egypt

Received 15 February 1999; accepted in revised form 31 August 1999

Key words: anodic behaviour, cast iron alloys, chloride ion, chromium, fluoride ion, nickel, phosphoric acid

Abstract

The anodic behaviour of four cast iron alloys containing up to 16.7% Ni, in deaerated 60 wt % H_3PO_4 with and without 5×10^{-3} M F^- , Cl^- ions and 1:1 Cl^-/F^- mixture was studied by the potentiostatic technique. Values of E_{corr} of the alloys are markedly influenced by their composition. The anodic behaviour in the active region is controlled by Fe in the alloys and the dissolution reaction is characterized by Tafel slopes, b_a , between 64 and 88 mV (decade) $^{-1}$. A two-electron transfer mechanism for the anodic dissolution is proposed. Passivation of the alloys is due to the formation of oxide layers including Fe_2O_3 and/or Fe_3O_4 . Both critical and passive c.d. (I_{cc} and I_p) are markedly increased in the presence of Cl^- ions, but the presence of F^- ions inhibit the active dissolution of the alloys. The Tafel slope for oxygen evolution reaction (o.e.r.) in the transpassive region, b_{O_2} is 240 ± 25 mV. In the proposed mechanism for the o.e.r., the rate determining step is an electron transfer reaction and possible interpretation of the high Tafel slopes is given based on the dual barrier model.

1. Introduction

The addition of Ni to cast iron promotes soundness and tends to reduce the size of graphite flakes; both factors favour uniform corrosion and the avoidance of localized attack and pitting. In many alloy cast irons the conjoint use of Ni and Cr gives very favourable results [1]. The early researches of Vanik and Merica [2] showed that addition of Ni (10–12%) to cast iron causes a moderate increase in the resistance to attack by 5% H_2SO_4 and further addition to ~18% causes rapid increase in resistance. Similar results were obtained by Everest and Hansen [3] with additions of 15–20% Ni. The passive behaviour of grey cast iron in 96 wt % H_2SO_4 was studied by Maahn [4]. He found that the material is normally passive in boiling concentrated acid and passivity is not possible with dilution to 93 wt % or the presence of reducing impurities. The anodic and cathodic behaviour of iron, different C-steels and cast iron in 0.1 M borax solution was studied by Tourky et al. [5]. They found that the anodic behaviour of the steels is markedly influenced by the nature of the separate phases in the Fe–C system. The amount of charge required for passivation was found to increase with C-content, attaining a maximum value with cast iron. The electrochemical and corrosion behaviour of three types of cast irons in acid sulphate solution of pH 1 was studied by Abdul-Azim and Sanad [6]. Their results indicated that the kinetic parameters (bc , i_o , E_{corr} and I_{corr}) depend

markedly on the amount and area of the separate phases. Suery and Hiltbrunner [7] found that additions of $\text{Ni} \leq 3\%$ to cast iron have no useful effect on the resistance to corrosion in both sea and fresh water. However, the corrosion rate of cast iron in solutions containing H_2S was found to decrease by 50% with small additions of Ni. It was reported [8] that the active dissolution of austenitic cast iron (20% Ni) occurs at more noble potentials than ferritic irons, and the value of i_{crit} decreases with increasing Cr and Si content. The anodic behaviour of two grey cast iron alloys I and II containing 0.51 and 0.92% Ni, respectively, in H_2SO_4 , HCl and NaOH solutions was studied by Hikmat et al. [9]. Their results revealed identical behaviour for the two alloys in H_2SO_4 and HCl solutions. In HNO_3 alloy I showed different behaviour from alloy II. The corrosion behaviour of various Cr-cast iron alloys in a quartzite slurry was investigated by Rajagopalan and Wasaki [10]. They found that high Cr-cast iron exhibited low corrosion rates at low and non abrasive conditions.

The aim of the present investigation is to study and compare the anodic behaviour of four cast iron alloys; two grey cast irons, 4.2 Ni-white cast iron and 3.4 Cr-austenitic cast iron, in relatively concentrated phosphoric acid solution (a major chemical product which has many important uses, especially in the fertilizer industry), and to study the effect of Cl^- , F^- ions and Cl^-/F^- mixture on such behaviour. The steady state potentiostatic technique was used.

2. Experimental details

2.1. Cast iron specimens

Annealed four cast iron alloys (C₋₁, C₋₂, C₋₃ and C₋₄) were used in this investigation. The compositions of the alloys are given in Table 1. Cylindrical specimens were machined 20 mm long and 5 mm in diameter. They were inserted in Teflon mounts in such a way that the flat surface was in contact with the solution.

2.2. Electrolytes

A 60 wt % solution of phosphoric acid (~9 M) was prepared from concentrated reagent grade acid and bidistilled water and was deaerated by passing O₂-free N₂ for 6 h before the electrode was introduced into the cell. N₂ bubbling was continued throughout the run. To determine the effect of Cl⁻ and F⁻ ions on the anodic behaviour of the examined alloys, appropriate volumes of concentrated solutions of NaCl and NaF were added to the cell.

2.3. Potential and polarization measurements

Prior to potential measurements, the specimens were mechanically polished with emery papers in steps down to grade 600, degreased with absolute ethanol (AR grade), washed with double distilled water and inserted in the cell which has been described elsewhere [11]. Stirring of the solution was achieved using a Gallenkamp magnetic stirrer and the stirring speed was kept constant during the experiment.

Potentials were measured with a Sargent–Welch electronic potentiometer, using a saturated calomel electrode (SCE) half cell. Potentials are quoted with respect to SCE at 30 °C.

Potentiostatic runs (anodic polarization) were carried out using a Wenking ST-72 potentiostat and a stepwise technique at two minute intervals. Starting at the steady corrosion potential, the potential was increased in the

noble direction using 20 mV steps in the active region up to the active-passive transition and 40 mV in the passive region. Each experiment was carried out at least twice.

3. Results and discussion

3.1. Open circuit corrosion potentials

The steady state corrosion potentials, E_{corr} , of the four cast iron alloys in deaerated 60% H₃PO₄ in the absence and the presence of 5×10^{-3} M Cl⁻, F⁻ and 1:1 F⁻/Cl⁻ mixture are given in Table 2. These data indicate that E_{corr} values for the alloys are clearly influenced by their composition. The most noble (more positive) potentials are those of the Cr-austenitic cast iron (alloy C₋₄).

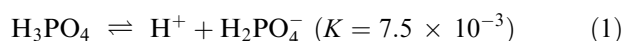
In the presence of 5×10^{-3} M F⁻ ion, E_{corr} values of all the alloys examined remained almost the same as in the pure acid solution. The results obtained in the presence of Cl⁻ ions (5×10^{-3} M) and the F⁻/Cl⁻ mixture show that E_{corr} values are not appreciably affected.

3.2.1. Active region and the mechanism of active dissolution

The anodic polarization curves of the alloys in 60% H₃PO₄ solution (pH ~ -1.2) are shown in Figure 1. Values of the anodic Tafel slope (b_a) obtained from the linear portion of the polarization curves in the active region were found to range from 64 to 88 mV/(decade)⁻¹. Values of b_a of ~60 mV (decade)⁻¹ were obtained by Condit [12] for the active region of dissolution for Fe, Fe-5Ni and Fe-10Ni in H₂SO₄ solution. He showed that Fe controls the active anodic behaviour of the Fe-Ni alloys up to 30% Ni.

Hackerman and McCafferty [13] reported values of b_a ranging from 60–75 mV (decade)⁻¹ for pure Fe in concentrated HCl solutions and admitted that such values are in accord with a two-electron mechanism of dissolution if $\alpha = 0.5$ to 0.4.

Because of the high concentration of H₃PO₄ (~9 M) and consequently the high concentration of H₂PO₄⁻ ion,



OH⁻ adsorption is not favoured and instead adsorption of H₂PO₄⁻ ions predominates. Thomas [14] found, during the passivation of Fe in neutral phosphate solutions, that at the potential where activity gives place

Table 1. Composition of cast iron alloys (in wt %)

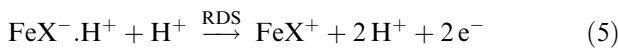
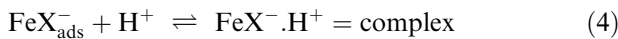
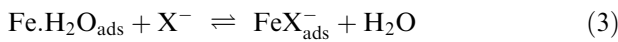
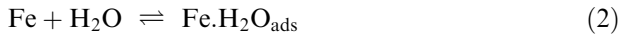
Cast iron alloys	C	Si	Ni	Cr
C ₋₁ (plain grey cast iron)	2.9	2.1	–	–
C ₋₂ (Ni grey cast iron)	3.6	1.8	2.6	–
C ₋₃ (Ni white cast iron)	3.9	0.37	4.2	–
C ₋₄ (Cr-austenitic cast iron)	3.4	2.7	16.7	3.4

Table 2. Corrosion potentials for cast iron alloys in deaerated 60% H₃PO₄ solution in presence of Cl⁻ and F⁻ ions

Specimen	$-E_{\text{corr}}$ vs SCE/mV			
	Pure medium	Cl ⁻ (5×10^{-3} M)	F ⁻ (5×10^{-3} M)	Cl ⁻ /F ⁻ (1:1)
Alloy C ₋₁	383	386	383	380
Alloy C ₋₂	367	367	366	365
Alloy C ₋₃	348	344	349	343
Alloy C ₋₄	290	285	288	285

to passivity the surface coverage with adsorbed phosphate ions is an appreciable fraction of a monolayer.

A two electron transfer mechanism for the anodic dissolution of the alloys, originating from that suggested by Hackerman and McCafferty [13] for pure Fe in 6 M HCl solution, can be proposed to account for the b_a values obtained;



where $\text{X}^- = \text{H}_2\text{PO}_4^-$.

This sequence is characterized by the parameter b_a :

$$b_a = \left(\frac{\partial E}{\partial \log ia} \right)_{a(\text{H}^+)a\text{X}^-} = \frac{2.303 RT}{F} = 60 \text{ mV} \quad (7)$$

($\alpha = 0.5$ and $T = 298 \text{ K}$)

Values of b_a ranging from 64 to 88 mV (decade) $^{-1}$ are in agreement with the proposed mechanism if $\alpha = 0.46$ to 0.34.

3.2.2. Behaviour in the passive region

The polarization curves shown in Figure 1 and the data given in Table 3, reveal that the anodic behaviour of both grey cast irons (alloys C_{-1} and C_{-2}) is almost the same. The primary passive potential (E_{pp}) for both alloys occurs at a value of -280 mV vs SCE, which agrees closely with the equilibrium potential of the system $\text{Fe}/\text{Fe}_3\text{O}_4$ ($E_{\text{Fe}/\text{Fe}_3\text{O}_4} = -256 \text{ mV}$ vs SCE at pH -1.2). The gradual development of passivation and the relatively high anodic currents in the active region suggest that the primary passivating layer is porous and has poor adherence to the substrate [15, 16]. This can be attributed to the graphite residue retained on the alloy surface during active dissolution. The results obtained by Flis [17] for iron-C alloys (containing up to 0.87% C), indicated that carbon retards passivation of iron in acidic phosphate solution (pH 4). The data given in Table 3, indicate that although both alloys C_{-1} and C_{-2} have the same value of E_{pp} , values of both critical and passive c.d.

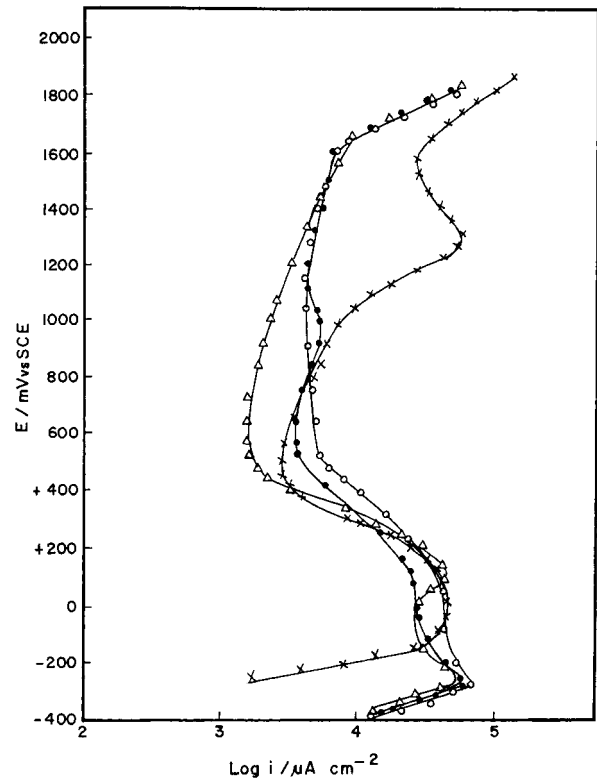


Fig. 1. Potentiostatic anodic polarization of Cast iron alloys in deaerated 60% H_3PO_4 solution. Alloy: (○) C_{-1} , (●) C_{-2} , (△) C_{-3} , (×) C_{-4} .

(I_{cc} and I_{p}) are lower in case of alloy C_{-2} (Ni grey cast iron) than alloy C_{-1} (plain grey cast iron). This is probably attributed to the presence of Ni, which was found to reduce the size of graphite flakes [8].

Typical SEM micrographs of the alloys are shown in Figure 2(a)–(d). In spite of the same etching depth, the alloys differ in surface morphology. The surface of alloy C_{-2} is uniformly covered with more fine graphite flakes than alloy C_{-1} .

The results obtained for alloy C_{-3} (Ni-white cast iron) (Figure 1 and Table 3) indicate that, although the value of E_{pp} is approximately the same as those of the alloys C_{-1} and C_{-2} , both I_{cc} and I_{p} are appreciably lower than those of alloy C_{-1} (I_{p} is about one-third that of alloy C_{-1}). On the other hand the anodic polarization curve manifests a second current peak (secondary passivation potential) at a value of $+80 \text{ mV}$ vs SCE which agrees closely with the equilibrium potential of the system $\text{Fe}_3\text{O}_4/\text{Fe}_2\text{O}_3$ ($E_{\text{Fe}_3\text{O}_4/\text{Fe}_2\text{O}_3} = +50 \text{ mV}$ vs SCE at

Table 3. Passivation parameters for cast iron alloys in deaerated 60 wt % phosphoric acid solution in presence of Cl^- and F^- and Cl^-/F^- mixture

Specimen Anion	Alloy C_{-1}			Alloy C_{-2}			Alloy C_{-3}			Alloy C_{-4}		
	E_{pp} /mV	I_{cc} mA /cm $^{-2}$	I_{p} mA /cm $^{-2}$	E_{pp} /mV	I_{cc} mA /cm $^{-2}$	I_{p} mA /cm $^{-2}$	E_{pp} /mV	I_{cc} mA /cm $^{-2}$	I_{p} mA /cm $^{-2}$	E_{pp} /mV	I_{cc} mA /cm $^{-2}$	I_{p} mA /cm $^{-2}$
Pure medium	-280	69.2	4.9	-280	64.6	3.7	-260	60.2	1.7	-20	52.5	3.1
$5 \times 10^{-3} \text{ M Cl}^-$	-260	72.4	15.1	-260	70.7	13.5	-220	69.0	15.8	-20	79.5	4.4
$5 \times 10^{-3} \text{ M F}^-$	-300	61.7	4.4	-280	52.5	3.3	-260	57.5	2.2	-40	42.7	3.0
Cl^-/F^- mixture (1:1)	-260	64.6	13.2	-260	63.1	12.0	-220	53.7	15.1	-20	66.2	5.7

pH -1.2). The relatively low value of I_p for the alloy indicates that the passive state of white cast iron containing Ni is much better than the passive state of grey cast iron. It was reported [18] that, owing to the lower Si content of white cast iron, Ni imparts a definite thickening influence on the matrix, thus counteracting porosity.

The results obtained for the alloy C₋₄ (Cr-austenitic cast iron) show different behaviour from that of the other alloys (SEM micrographs for the alloy, Figure 2(d), reveal that many spheroidal carbide particles are randomly dispersed throughout the austenitic matrix). Although values of I_{cc} and I_p are markedly lower than those of the grey cast irons, E_{pp} is 260 mV positive than the corresponding ones for such alloys. This value is ~ 70 mV more negative than the equilibrium potential of the system Fe_3O_4/Fe_2O_3 at the same solution pH. This can be attributed to the presence of some NiO in the passivating film ($E_{Ni/NiO} \approx -190$ mV at pH -1.2).

It was suggested by several authors [19–21] that the passivating film on iron is composed of an inner layer of Fe_3O_4 and an outer layer of $\gamma-Fe_2O_3$. The anodic polarization curve also reveals that the stable passive region covers a relatively narrow potential range (~ 100 mV). At potentials >550 mV, I_p increases with potential up to the commencement of a pseudo transpassive region, beyond which a secondary passivation potential is exhibited by the curve at a value of about $+1280$ mV. This value is comparable with the equilibrium potential of the system Ni_2O_3/NiO_2 at the same solution pH ($E_{Ni_2O_3/NiO_2} = +1232$ mV vs SCE at pH -1.2). Similar results were obtained by Sayano and Nobe for Ni in 0.5 M H_2SO_4 [22].

3.2.3. Transpassive region and the mechanism of the oxygen evolution reaction

All alloys exhibited transpassive behaviour at about $+1600$ mV vs SCE which is attributed to oxygen evolution. Condit [12] reported values of $+1500$ mV

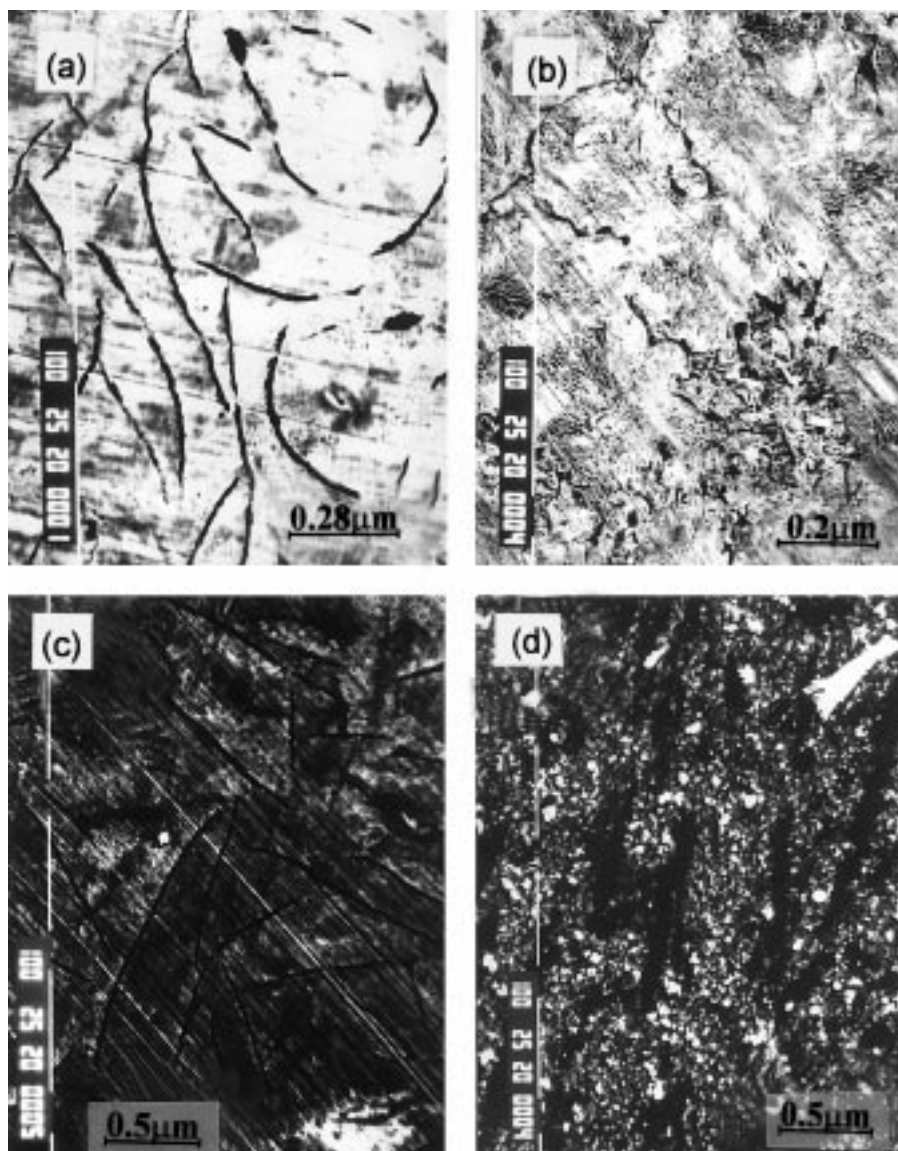
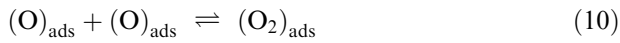
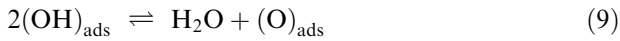
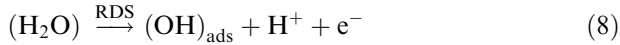


Fig. 2. SEM micrographs of specimens subjected to nitric acid etching. Alloy: (a) C₋₁ (350 \times), (b) C₋₂ (500 \times), (c) C₋₃ (200 \times) and (d) C₋₄ (200 \times).

for the transpassive region for Fe and some Fe–Ni alloys in 0.5 M H_2SO_4 . Values of the Tafel slope for o.e.r. (b_{O_2}) obtained directly from the $E/\log I$ curves in the transpassive region amount to 215 mV for the alloys C_{-1} , C_{-2} and C_{-3} and 265 mV for the alloy C_{-4} .

Values of $b_{\text{O}_2} = 220 \pm 10$ mV for o.e.r. on a PbO_2 electrode were obtained by Abdel-Aal et al. [23].

Assuming that the rate determining step in the o.e.r. is an electron transfer reaction probably occurring during the discharge of H_2O molecules from acid solutions [24], then the mechanism can be represented as



This sequence is characterized by the following parameter:

$$b_a = \left(\frac{\partial E}{\partial \log I_a} \right)_{a(\text{H}^+)} = \frac{2.303 RT}{\alpha F} = 120 \text{ mV} \quad (\alpha = 0.5 \text{ and } T = 298 \text{ K}) \quad (12)$$

Values of $b_{\text{O}_2} = 240 \pm 25$ mV (decade) $^{-1}$ in the present study indicate that charge transfer can not take place in such a simple manner.

It has been reported by several authors [14, 17, 25] that the passivating film formed on Fe, Fe–C and austenitic stainless steel in neutral and acidic phosphate solutions contain inclusions of iron phosphate. According to Dabosi et al. [26] inclusions of iron phosphate in the passivating iron oxide film are accompanied by a decrease in conductivity of the film.

The high Tafel slope of the o.e.r. may be interpreted on the basis of the dual barrier model reported firstly by Mayer [27] and applied by Conway and Vijn [28] and Lorenz et al. [29] for explaining anomalously high Tafel slopes. Thus, it is reasonable to assume that electrons of the charge transfer reaction have to surmount two energy barriers, the first corresponding to the passivating film and the second to the ionic double layer.

According to this treatment, the overall transfer coefficient α_t is given for the o.e.r. by

$$\alpha_t = \frac{\alpha_1 \alpha_2}{\alpha_1 + \alpha_2} \quad (13)$$

Assuming $\alpha_1 = \alpha_2 = 0.5$, then $\alpha_t = 0.25$, and

$$b_{\text{O}_2} = \frac{2.303 RT}{\alpha_t F} = 240 \text{ mV (decade)}^{-1} \text{ at } T = 298 \text{ K} \quad (14)$$

which is in agreement with the experimental results.

3.2.4. Effect of Cl^- and F^- ions and Cl^-/F^- mixture

The data given in Table 3 and Figures 3 and 4 show that Cl^- ions at a concentration of 5×10^{-3} M increases both I_{cc} and I_p for all the investigated alloys and shift E_{pp} by 20–40 mV in the positive direction. This suggests that Cl^- ions (at such concentration) accelerate the anodic dissolution of the alloys in both active and passive regions. The large increase in I_p for the alloys C_{-1} , C_{-2} and C_{-3} (I_p is 3–8 times the original value) denotes attack of the passivating oxide film by Cl^- ions resulting in a very porous film [21]. The relatively small increase in I_p for the alloy C_{-4} ($\sim 50\%$ increase) can be attributed to the Cr-content in the mixed oxide [30].

The presence of F^- ions (5×10^{-3} M) markedly decreases the value of I_{cc} for all the alloys, slightly affects their I_p values and increases the stability of the passive state. These results are in agreement with those obtained by Hart [31] and by Farina et al. [32] for some stainless and Ni base alloys in about 75% H_3PO_4 . It was reported by Schorr [33] that the formation of a stable monofluoro phosphoric acid (complexing F^- ions) reduces its corrosivity. It may be assumed that adsorption of $\text{HF-H}_3\text{PO}_4$ complex results in inhibition of the active dissolution of the alloys and stabilizes their passive state. Adsorption of some complex anions [34] and cations [35] was found to play an important role in mild steel inhibition in some carboxylic acids and NaCl solutions.

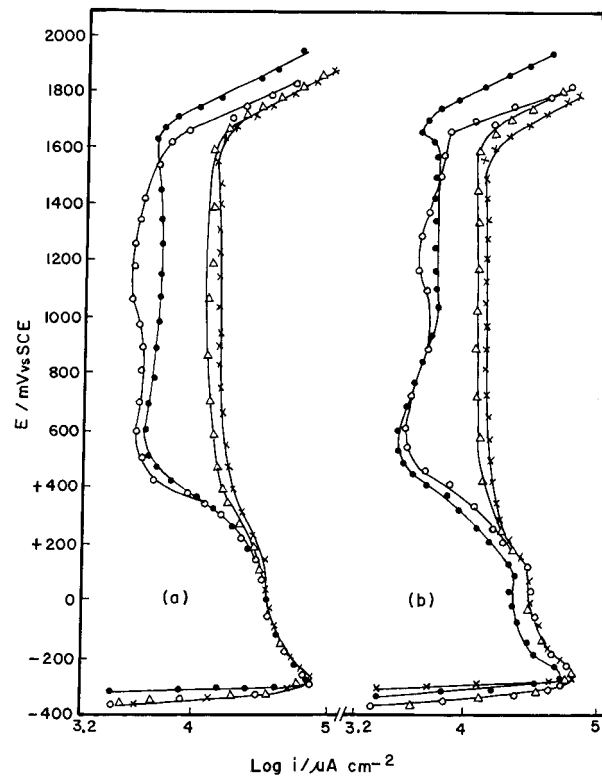


Fig. 3. Potentiostatic anodic polarization of alloys: (a) C_{-1} , (b) C_{-2} in deaerated 60% H_3PO_4 solution in presence of Cl^- and F^- ions. Key: (○) pure medium (×) 5×10^{-3} M Cl^- , (Δ) 1:1 Cl^-/F^- mixture, (●) 5×10^{-3} M F^- .

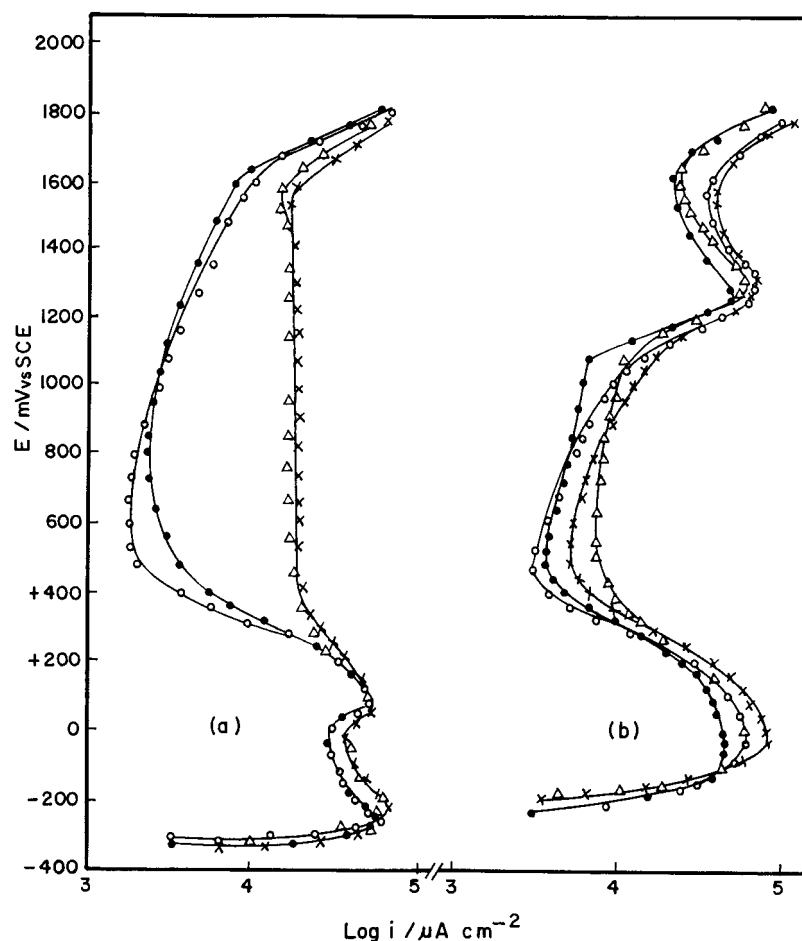


Fig. 4. Potentiostatic anodic polarization of alloys: (a) C-3, (b) C-4 in deaerated 60% H_3PO_4 solution in presence of Cl^- and F^- ions, (○) pure medium, (×) 5×10^{-3} M Cl^- , (Δ) 1:1 Cl^-/F^- mixture, (●) 5×10^{-3} M F^- .

Although addition of F^- ions in combination with Cl^- ions (1:1) considerably decreases I_{cc} values for all the alloys compared with those obtained in solutions containing Cl^- ions alone, I_{p} values are slightly affected. This suggests that F^- ions (in the complexed form) can inhibit Cl^- ion attack in the active region, but do not exert a pronounced effect on such attack in the passive region.

4. Conclusions

- (i) In deaerated 60 wt % phosphoric acid, the corrosion potentials of all the examined alloys were found to be markedly influenced by their composition and the most noble value was that of Cr-austenitic cast iron.
- (ii) Iron controls the active dissolution of the alloys irrespective of the Cr and/or Ni content. Tafel slopes (b_a) of the active regions, ranging from 64 to 88 mV (decade) $^{-1}$ are in accord with a two-electron transfer (60 mV (decade) $^{-1}$) mechanism of dissolution if $\alpha = 0.47$ to 0.34.
- (iii) The Cr-austenitic cast iron has the greatest tendency to passivate and the Ni-white cast iron

has the highest degree of passivation (lower value of I_{p}).

- (iv) At a concentration of 5×10^{-3} M, Cl^- ion enhanced the anodic dissolution of the alloys in both the active and passive regions, while F^- ion at the same concentration, inhibited active dissolution.
- (v) All alloys exhibited transpassive behaviour at approximately the same potentials. In the proposed mechanism for oxygen evolution, the rate determining step is an electron transfer reaction during the discharge of H_2O molecules from the acid solution. Electrons have to surmount two energy barriers, the first corresponding to the passivating film and the second to the ionic double layer.

References

1. A.M. Hall, 'Nickel in Iron and Steel' (Wiley & Sons, New York, 1954, 255).
2. J.S. Vanick and P.D. Merica, *Trans. Amer. Soc. (Steel treating)* **18** (1930) 923.
3. A.B. Everest, T.H. Turner and D. Hanson, *J. Iron Steel Inst.* **116** (1927) 185.
4. E. Maahn, *Br. Corros. J.* **1** (1966) 350.
5. A.R. Tourky, A.A. Abdul-Azim and S.H. Sanad, *Corros. Sci.* **8** (1968) 857.

6. A.A. Abdul-Azim and S.H. Sanad, *Corros. Sci.* **12** (1978) 337.
7. P. Suery and K. Hiltbruner, *Arch. Eisen Huetten Wes.* **50** (1979) 493.
8. L.L. Shrier (ed.), 'Corrosion of high Ni Cast Iron' Vol. 1 (Newens-Butterworth, London, 1978, 3, p. 100 and p. 393).
9. L.M. Al-Shama, J.M. Saleh and N.A. Hikmat, *Corros. Sci.* **27** (1987) 221.
10. V. Rajagopal and I. Wasaki, *Corrosion* **48** (1992) 124.
11. M.S. Abdel-Aal, I.M. Issa and A. Zedan, *J. Appl. Electrochem.* **6** (1976) 79.
12. D.O. Condit, *Corros. Sci.* **12** (1972) 451.
13. E. McCafferty and N. Hackerman, *J. Electrochem. Soc.* **119** (1972) 999.
14. J.G.N. Thomas, *Br. Corros. J.* **1** (1966) 156.
15. J.W. Johnson, Y.C. Sunn and N.J. James, *Corros. Sci.* **11** (1971) 53.
16. J. Parkins, W.H. Lucbke, K.J. Grahame and J.M. Tudd, *J. Electrochem. Soc.* **124** (1977) 819.
17. J. Flis, *Corros. Sci.* **25** (1985) 317.
18. E.C. Piggot, 'Ferrous Analysis, Modern Practice and Theory', 2nd edn (Chapman & Hall, London, 1954), p. 292.
19. G.L. Foley, J. Kruger and C.J. Bechtold, *J. Electrochem. Soc.* **114** (1967) 936.
20. M. Cohen, *J. Electrochem. Soc.* **121** (1974) 191c.
21. D.O. Condit, *Corrosion* **28** (1972) 95.
22. R.R. Sayano and Ken Nobe, *Corrosion* **22** (1966) 81.
23. I.M. Issa, M.S. Abdel-Aal and A.A. El-Miligy, *J. Appl. Electrochem.* **5** (1976) 27.
24. J.P. Farr and N.A. Hampson, *Chem. Rev.* **72** (1972), 702.
25. A. Guenbour, J. Faucheu and A. Ben Bachir, *Corrosion* **44** (1988), 214.
26. A. Guenbour, N. Bui, J. Faucheu, Y. Segui, A. Ben Bachir and F. Dabosi, *Corros. Sci.* **30** (1990) 189.
27. R.E. Mayer, *J. Electrochem Soc.* **107** (1960) 847.
28. B.E. Conway and A.K. Vijh, *J. Phys. Chem.* **71** (1967) 3637.
29. M.S. Abdel-Aal, A.A. El-Miligy, G. Reiners and W.J. Lorenz, *Electrochim. Acta* **20** (1975) 507.
30. S. El-Hajjaji, J. Roy, L. Aries and F. Dabosi, *Br. Corros. J.* **28** (1993) 201.
31. A.C. Hart, *Br. Corros. J.* **6** (1971) 205.
32. P. Baracco, G. Chiaro and A. Farina, Proc. 7th European Symposium on 'Corrosion Inhibitors', Ann. Univ. Ferrara N.S. Sez. V Suppl. No. 9 (1990), p. 1341.
33. A. Alon, J. Yahalom and M. Schorr, *Corrosion* **31** (1975), 315.
34. M.S. Abdel-Aal and M.H. Wahdan, *Br. Corros. J.* **23** (1988), 25.
35. C. Wrubi, E.D. Mor and U. Montini, Proceedings of the 6th European Symposium on 'Corrosion Inhibitors', Ann. Univ. Ferrara N.S. Sez. V Suppl. No. 8 (1985), p. 557.